

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of:

Group Art Unit: 1796

ANDREAS BACHER, et al.

Examiner: Vickey M. Ronesi

Serial No.: 10/618,936

Filed: July 14, 2003

For: SILANE-CONTAINING POLYVINYL ALCOHOL FOR COATING  
SLIPS

Attorney Docket No.: WAS 0595 PUS

**DECLARATION OF DR. ANDREAS BACHER**

Mail Stop Amendment  
Commissioner for Patents  
U.S. Patent & Trademark Office  
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Sir:

I, Dr. Andreas Bacher, do hereby declare and state as follows:

1. I am a co-inventor of the claimed subject matter of U.S. application Serial No. 10/618,936 filed July 14, 2003.

2. I obtained my Ph.D. in Macromolecular Chemistry from the University of Bayreuth, Germany in 1998. In 1998-99, I pursued post doctoral research at the University of Sheffield, England, and since January 1 of 2000 I have been employed by the assignee, Wacker Chemie AG. During my employment at Wacker Chemie, my research and development work was first in the field of polyvinyl alcohols, particularly modified and functionalized polyvinyl alcohols, in emulsions, dispersions, and dispersible polymer powders for the construction and coatings markets, and in organosilicone nanoparticles. Since 2003, my research and development work is for the Silicone Division, particularly in the technical field of silicone rubber. I am the named inventor or co-inventor of numerous patents and their equivalents worldwide, including: U.S. 6,573,326; EP 1180535 B1; EP 1180529 B1; EP 1258464 B1; EP 1253157 A1; EP 1549682 B1; EP 1530595 B1; DE 10232664; U.S. 7,052,773; EP 1382621 B1;

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DE 10320629; EP 1625177 B1; EP 1625254 B1; EP 1620271 B1; EP 1593721 A1; EP 1812478 A1; EP 1819742 A1.

3. I am familiar with the present application, its prosecution history, the recent Office Action, and the prior art.

4. In my opinion, based on my experience with polyvinyl alcohols and modified polyvinyl alcohols, one skilled in the art would not be motivated to prepare a polyvinyl alcohol copolymer also containing, in addition to vinyl acetate-derived vinyl alcohol moieties, moieties derived from isopropenyl acetate (1-methylvinyl acetate) and unsaturated silane monomers, based on the teachings of Schilling and Maruyama.

5. Schilling desired to produce a polyvinyl alcohol copolymer which minimized pigment shock in producing coating slips and other highly pigment-loaded dispersions. To do so, Schilling incorporated isopropenyl acetate into his copolymer mixture, thus producing polymer containing isopropenyl alcohol units in addition to vinyl alcohol units. However, the use of isopropenyl acetate also involves a considerable cost penalty, since isopropenyl acetate is considerably more expensive than vinyl acetate, the monomer from which polyvinyl alcohols are generally prepared. Unless faced with some reason for its use, one skilled in the art would not employ isopropenyl acetate.

6. Maruyama discloses silane-modified polyvinyl alcohols prepared, in one method, by copolymerizing vinyl acetate and an unsaturated silane. The Maruyama copolymers are not known in the industry to exhibit marked pigment shock as are polyvinyl alcohols. A possible reason for this is that all the repeating moieties of polyvinyl alcohol, having pendant OH groups, are highly hydrophilic and offer the possibility of a great deal of hydrogen bonding with pigment surfaces, whereas in silane-modified polyvinyl alcohols, the regularity of repeating vinyl alcohol units is disrupted by the presence of random silane units, which also render these polymers less hydrophilic.

However, the silane-modified copolymers of Maruyama are known to suffer from viscosity stability in aqueous solution. This is shown in the present application by

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the Comparative Example, wherein the viscosity of a Maruyama-type silane-modified polyvinyl alcohol, POVAL® R-1130, increased by a factor of more than 3 over storage of just 2 weeks.

In fact, one skilled in the art, reading the Maruyama specification, immediately recognizes that Maruyama desires rapid reaction of the silicon-bound alkoxy groups so that the Maruyama alkoxy silane-modified polyvinyl alcohol quickly reacts with cellulosic fibers, forming a gelled coating on the surface, which prevents further absorption of the coating composition into the paper. However, in an aqueous dispersion, the same high alkoxy-group reactivity is also responsible for the viscosity increase upon storage, due to chain extension and crosslinking through these alkoxy groups. Thus, based on Maruyama's teachings, one skilled in the art would be dissuaded from further modifying his silane-modified polyvinyl alcohols to be less reactive; rather one would wish them to be more reactive to provide even less penetration into the substrate. Applicants have proceeded in a direction opposite to the teachings of Maruyama, and yet achieved not only a more stable polymer, but produced higher abrasion resistance at the same time.

7. Schilling does not teach or suggest how to modify the polymers of Maruyama to solve their stability problem. Moreover, no one skilled in the art would be motivated to modify a Schilling isopropenyl alcohol moiety-containing polyvinyl alcohol, by incorporating an unsaturated silane into the polymerizable monomer mixture as taught by Maruyama. Prior to Applicants' invention, it was not known that incorporation of isopropenyl alcohol moieties into a silane-modified polyvinyl alcohol would increase its storage stability. This novel and useful feature was discovered by Applicants. Since the silane-modified polyvinyl alcohols are not known to suffer from pigment shock, there would be no reason to add the expensive isopropenyl acetate comonomer to solve a "problem" which does not even exist.

If Schilling, for example, were motivated to employ a silane-functional comonomer, there would be no reason apparent to Schilling, or more generally to anyone skilled in the art, to also employ isopropenyl acetate as a comonomer. Why would one do this? Unsaturated silane comonomers are themselves expensive. Including isopropenyl acetate as a comonomer would only increase cost without any known or perceived benefit.

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8. The Examples and Comparative Examples illustrate the benefits of the subject invention. It was desired to increase abrasion resistance of coatings employing silane-modified polyvinyl alcohols without compromising the remaining properties and while retaining ease of production, including storage stability. See, e.g., page 9 of the Specification, last paragraph.

These objects are accomplished by including isopropenyl acetate as well as an unsaturated silane as comonomers with vinyl acetate to produce, by subsequent hydrolysis, a modified polyvinyl alcohol terpolymer. These polymers not only exhibited higher abrasion resistance than the polymers of Kuraray (Maruyama), but also exhibited simply outstanding viscosity stability. Both these results are completely surprising and unexpected based on the teachings of Schilling and Maruyama, whether alone or in combination.

9. The showing is believed to be commensurate with the present scope of the claims. In particular, the Examples and Comparative Examples are squarely within relatively narrow compositional ranges of the claimed terpolymers, and there is no scientific reason to expect that the same results would not be obtained throughout the claimed ranges.

10. To augment the previous showing, Example 1 was repeated with a different silane, vinylmethyldiethoxysilane, and compared with former Example 1 and Comparative Example 1. The makeup of the Kuraray POVAL® R-1130 is also set forth below. The Experiments were performed in the assignee's laboratories in Burghausen, Germany, as follows:

#### Example 1

612 g of water, 61.2 mg of copper(II) acetate and 61.2 g of a 5% strength polyvinylpyrrolidone solution (PVD-K90) in water were initially introduced under nitrogen into a thermostated laboratory apparatus having a capacity of 2.5 liters. A solution of 620 mg of tert-butyl per-2-ethylhexanoate (TBPEH 99% strength in water), 322 mg of tert-butyl pernonodecanoate (Pergan PND 95% strength in water), 4.9 g vinyltriethoxysilane, 48.9 g of isopropenyl acetate and 42.8 g of isopropanol in 612 g of vinyl acetate were run in while stirring. The reactor was heated to 51.5°C and, after the reaction had diminished, was heated stepwise to

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75°C. This temperature was maintained for a further 2 hours, after which cooling was carried out. The resulting polymer beads were filtered off with suction, washed thoroughly with water and dried.

In a laboratory reactor having a capacity of 2.5 liters, 90 g of polymer beads were dissolved in 810 g of methanol at 50°C. The solution was cooled to 30°C, a layer of 500 g of methanol was introduced on top with the stirrer stationary, and the methanolic NaOH was immediately added (10 g of NaOH, 46% strength in water, dissolved in 90 g of methanol) and the stirrer was switched on.

The solution became increasingly turbid. During the gel phase, the stirrer was set to a higher speed in order to comminute the gel. After the gel phase, the reaction was allowed to continue for a further 2 hours, neutralization was effected with acetic acid and the solid form was filtered off, washed and dried. A completely hydrolyzed polyvinyl alcohol having a Höppler viscosity of 28 mPas (4% strength in water) was obtained.

#### Example 2

Example 2 was performed analogously to Example 1, with the difference that 5.0 g vinylmethyldiethoxysilane were applied instead of vinyltriethoxysilane. The respective hydrolyzed polyvinyl alcohol had a Höppler viscosity of 26 mPas (4% strength in water).

#### Comparative Example 1:

Commercial silane-containing polyvinyl alcohol (Poval R-1130 from Kuraray) from vinylacetate and 0.55 weight percent vinyltrimethoxysilane, based on total polymer, having a Höppler viscosity of 25 mPas (4% strength in water).

Testing of performance characteristics:

The polyvinyl alcohols from the example and comparative example were tested in the following formulations:

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**Formulation of coating slip for paper coating:**

Precipitated silica	100 parts by weight
Polyvinyl alcohol	35 parts by weight
Cationic dispersant	5 parts by weight
Solids content of the coating slip:	20 - 35% by weight

The coating slip was tested on base paper: sized paper, 80 g/m<sup>2</sup>. The coat was 15 g/m<sup>2</sup>.

**Formulation for coating slip for coating plastics foils:**

Precipitated silica	100 parts by weight
Polyvinyl alcohol	34 parts by weight
Cationic dispersant	8 parts by weight
Polymer dispersion	50 parts by weight
Solids content of the coating slip:	20 - 28% by weight

The coating slip was tested on a 90 g/m<sup>2</sup> PET foil provided with an adhesion promoter. The coat was 15 g/m<sup>2</sup>.

**Abrasion test:**

A 4.5 cm wide and 19 cm long coated paper strip or foil strip was processed by means of 50 strokes in an abrasion tester from Prüfbau (Dr. Dürner system) with a black drawing paper applied to a stamp (500 g).

The resulting black paper was then assessed visually, the rating 1 representing the optimum.

**Brightness:**

The brightness was determined by means of reflectance measurement using a R 457 filter and was visually assessed, the rating 1 representing the optimum.

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**Storage stability:**

The storage stability was tested by storing in each case 11% strength aqueous solutions of the polyvinyl alcohols at a pH of 8.5 for up to 14 days and determining the Höppler viscosity (according to DIN 53015, as 4% strength by weight aqueous solution).

Table 1 (Results on paper):

Coat analysis	Example 1	Comparative Example 1	Example 2
Abrasion test	1.0	1.5	1.2
Brightness visual	1.0	1.0	1.0
Brightness R457	5.1	5.2	5.1

Table 2 (Results on plastics foil):

Coat analysis	Example 1	Comparative Example 1	Example 2
Abrasion test	1.0	1.5	1.1
Brightness visual	1.0	1.0	1.0
Brightness R457	5.3	5.3	5.3

Table 3 (Shelf life):

Storage time	Höppler viscosity [mPas]		
	Example 1	Comparative Example 1	Example 2
immediately	28.1	25.1	26.0
1 day	28.1	27.8	26.0
2 days	28.1	31.4	26.0
3 days	28.2	35.8	26.0
5 days	28.2	42.9	26.1
7 days	28.2	61.7	26.1
14 days	28.3	90.5	26.1

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11. As can be seen, changing the silane from that of Example 1 did not change the terpolymer stability. Both the terpolymer of Example 1 and that of Example 2 exhibited excellent stability, whereas a copolymer of Kuraray exhibited a very considerable viscosity increase. These results are truly surprising and unexpected. No one skilled in the art would have expected that a terpolymer containing vinyl alcohol, isopropenyl alcohol, and silane moieties would either be storage stable or produce the increase in abrasion resistance obtained.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code.

  
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DR. ANDREAS BACHER

Dated: 05/17/08